NEUTRON ACTIVATION ANALYSIS BY CADMIUM RATIO AND PAIR COMPARATOR METHODS

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الخلاصة :

في هذه الدراسة قدمت طريقتين جديدتين عياريتين طورت للتحليل التنشيطي للنيوترونات .

تم تعيين معاملات التنشيط بعد تجهيز العينات بصورة تجعل من قيمة الامتصاص الذاتي كمية مهملة تماماً في نفس الوقت تم إيجاد العلاقة بين معاملات التنشيط لكل من حالتي التنشيط الحراري وفوق الحراري ، والتي كانت عبارة عن متوسط عدة قياسات لكل نظير على حدة .

أهم ميزة غير عادية في هذه الطرق هو علاجها لتأثير تغير طيف النيوترونات عند مناطق التشعيع المختلفة على معاملات التنشيط الفعالة .

وللتأكد من كفاءة تلك الطرق ، طبقت لتعيين تركيز العناصر قليلة التركيز في عينات جيولوجية عيارية من صخور الجرانيت G–1 وكانت النتائج متفقة بصورة جيدة مع النتائج المنشورة .

ABSTRACT

New standardizations have been developed for neutron activation analysis. In these methods, experimental activation factors are determined. The unusual feature of these techniques is a solution for the effects on the effective activation factors resulted from the variation of neutron spectrum at different irradiation positions. The methods were used to determine the concentrations of trace elements in USGS standard rocks G–2; the results were in agreement with literature values.

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INTRODUCTION

It is well known that the effective activation factors is in close relationship with the *b* neutron spectrum when neutron activation analysis is applied by the single comparator method (SCM) [1, 2]. In this paper new methods denoted as 'cadmium ratio method' and 'pair comparator method' are described, which can be used for activation analysis with a research reactor and provide a solution for the effects on the effective activation factors from the variations of neutron spectrum at different irradiation positions which are usually encountered in the SCM.

THEORETICAL FOUNDATION

When a sample is irradiated and measured, the photopeak counting rate of a certain nuclide of interest in the sample leads to the determination of the weight of the corresponding element:

$$A^0 = Km \tag{1}$$

$$= N_p/(1-e^{-\lambda t_{irr}})(e^{-\lambda t_d})\, C\epsilon_p \tag{2}$$

$$\text{when } K = \frac{6.02 \times 10^{23}}{M}\theta\hat{\sigma}\phi\gamma \tag{3}$$

and

m = weight of the element

M = atomic weight of the element

N_p = counting rate of the nuclide at the beginning of the measurement

C = constant depending on the source-to-detector geometry

ϕ = neutron flux $= \phi_{th} + \phi_{ep}$; where ϕ_{th} is the thermal neutron flux and ϕ_{ep} is the epithermal neutron flux.

$\hat{\sigma}$ = effective activation cross-section.

$$= \sigma_{th}\frac{\phi_{th}}{\phi_{th}+\phi_{epi}}+I_0\frac{\phi_{epi}}{\phi_{th}+\phi_{epi}} \text{ where}$$

σ_{th} is the thermal activation cross-section and I_0 is the resonance integral.

γ = abundance of gamma-ray.

θ = natural abundance of the target nuclide,

ϵ_p = intrinsic photopeak efficiency of the detector for a particular gamma-ray,

λ = disintegration constant of the product nuclide,

t_{irr} = irradiated period.

t_d = cooling period.

SINGLE COMPARATOR METHOD

When an element 1 is irradiated with a 'comparator' 2, the following simple relation holds

$$\frac{A_1^0}{A_2^0}=K_{1/2}\frac{m_1}{m_2} \tag{4}$$

but

$$K_{1/2}=\frac{M_2\theta_1\gamma_1\hat{\sigma}_1}{M_1\theta_2\gamma_2\hat{\sigma}_2} \tag{5}$$

$$K_{1/2}=K_0\frac{1+\left[\dfrac{I_0}{\phi_{th}}\right]_1\left[\dfrac{\phi_{epi}}{\phi_{th}}\right]}{1+\left[\dfrac{I_0}{\phi_{th}}\right]_2\left[\dfrac{\phi_{epi}}{\phi_{th}}\right]} \tag{6}$$

where

$$K_0=\frac{M_2\theta_1\gamma_1\sigma_{th,1}}{M_1\theta_2\gamma_2\sigma_{th,2}}$$

If $K_{1/2}$ values are available for the element 1, the concentration for it can be calculated simply as

$$\rho_{1(ppm)}=\frac{m_2\,(\mu g)}{m_s\,(g)}\frac{A_1^0/A_2^0}{K_{1/2}}$$

Where m_s represents the sample weight in grams and m_2 represents the comparator in micrograms. But because $K_{1/2}$ dependends on the neutron spectrum, as shown in Equation (6), a definite value of $K_{1/2}$ can be used only for the same neutron spectrum at the same position of irradiation. This is the main drawback of this method.

THE CADMIUM RATIO METHOD

The cadmium ratio R_{Cd} of a nuclide is defined as the ratio of the specific count rates obtained after irradiation of a comparator with and without cadmium cover. This, the Cd-ratio can be written as:

$$R_{Cd}=\frac{A_{sp}}{(A_{sp})_{Cd}}=\frac{\alpha_{th}\phi_{th}+I_0\phi_{ep}}{I_0\phi_{ep}}$$

$$= \frac{\phi_{th}}{\phi_{ep}} \frac{\sigma_{th}}{I_0} + 1 \qquad (7)$$

where

A_{sp}: Specific count ratio $= \theta \times 6.02 \times 10^{23}$

$$\frac{(\phi_{th}\sigma_{th} + \phi_{epi}I_0)\gamma\epsilon_{p.c}}{}$$

$(A_{sp})_{Cd} \qquad = \theta \times 6.02 \times 10^{23}$

$$\phi_{eI_0}\gamma\epsilon_{p.c}$$

If $F = \phi_e/\phi_{th}$, $Q_0 = I_0/\sigma_{th}$

Equations (6) and (7) can be expressed as:

$$K_{1/2} = K_0 \cdot \frac{1 + Q_0, 1 F}{1 + Q_0, 2 F}$$

$$= K_0 \cdot \frac{\dfrac{1}{Q_0, 2 F} + \dfrac{Q_0, 1}{Q_0, 2}}{\dfrac{1}{Q_0, 2 F} + 1} \qquad (8)$$

$$R_{Cd2} = \frac{1}{F Q_0} + 1. \qquad (9)$$

From Equation (9)

$$\frac{1}{Q_0, 2 F} = R_{Cd2} - 1 \qquad (10)$$

Then Equation (8) becomes

$$K_{1/2} = K_0 \left(\frac{R_{Cd2} - 1 + \dfrac{Q_0, 1}{Q_0, 2}}{R_{Cd2}} \right)$$

$$= K_0 - \left(K_0 - \frac{Q_0, 1}{Q_0, 2} K_0 \right) / R_{Cd2}$$

$$= K_0 - (K_0 - K_{e,0}) / R_{Cd2} \qquad (11)$$

where

$$K_{e,0} = \frac{Q_0, 1}{Q_0, 2} K_0 \qquad (12)$$

From Equation (11) it is shown, that the activation factor $K_{1/2}$ is dependent on the cadmium ratio R_{Cd2}

where $0 \le \dfrac{1}{R_{Cd2}} \le 1$

If $\dfrac{1}{R_{Cd2}} = 0$, $K_{1/2} = K_0$

where K_0: thermal neutron activation factor

While $\dfrac{1}{R_{Cd2}} = 1$, $K_{1/2} = K_{e,0}$

where $K_{e,0}$: epithermal neutron activation factor.

From the linear relationship between $K_{1/2}$ and $1/R_{Cd2}$ or from the values of K_0 and $K_{e,0}$ the value of $K_{1/2}$ at any irradiated position can be calculated.

PAIR COMPARATOR METHOD

In this method an element '1' is irradiated with the comparators '2' and '3' simultaneously; then the following relation holds:

$$\frac{A_1^0}{A_2^0} = K_{1/2} \frac{m_1}{m_2}; \quad \frac{A_3^0}{A_2^0} = K_{3/2} \frac{m_3}{m_2}.$$

From Equation (8)

$$K_{1/2} = K_0 \frac{1 + Q_0, 1F}{1 + Q_0, 2F}$$

$$= K_0 \frac{1 + (Q_0, 1/Q_0, 2)(Q_0, 2F + 1) - (Q_0, 1/Q_0, 2)}{1 + Q_0, 2 F}$$

$$= \frac{K_0(1 - Q_0, 1/Q_0, 2)}{1 + Q_0, 2F} + \left(\frac{Q_0, 1}{Q_0, 2} \right) K_0. \qquad (13)$$

According to Equation (13), the $K_{3/2}$ factor can be expressed as:

$$K_{3/2} = \frac{K_{0(3/2)}(1 - Q_{0,3}/Q_{0,2})}{1 + Q_{0,2}F}$$

$$+ K_{0(3/2)}(Q_{0,3}/Q_{0,2}). \qquad (14)$$

From Equations (13) and (14), $Q_{0,2}F$ can be eliminated:

$$\therefore K_{1/2} = K_{0(1/3)} \frac{Q_{0,1} - Q_{0,2}}{Q_{0,3} - Q_{0,2}} K_{3/2}$$

$$= K_0 \frac{Q_{0,1} - Q_{0,3}}{Q_{0,2} - Q_{0,3}}. \qquad (15)$$

From Equation (15) $K_{1/2}$ can be calculated by using the value of $K_{3/2}$ and it is shown clearly that the ratios $K_{1/2}/K_{1/3}$ are independent of special distribution and allow measurements to be made in a variety of circumstances.

EXPERIMENTAL

All K factors were experimentally determined and calculated versus the 411.974 keV γ-line of [198]Au and [95]Zr–[97]Zr. K-factors were obtained as an average of at least three determinations. In every irradiation, the comparator and the element to be investigated were packed close together in polyethylene vials or in a cylindrical Cd box (1 mm wall thickness, $h/d = 2$). The
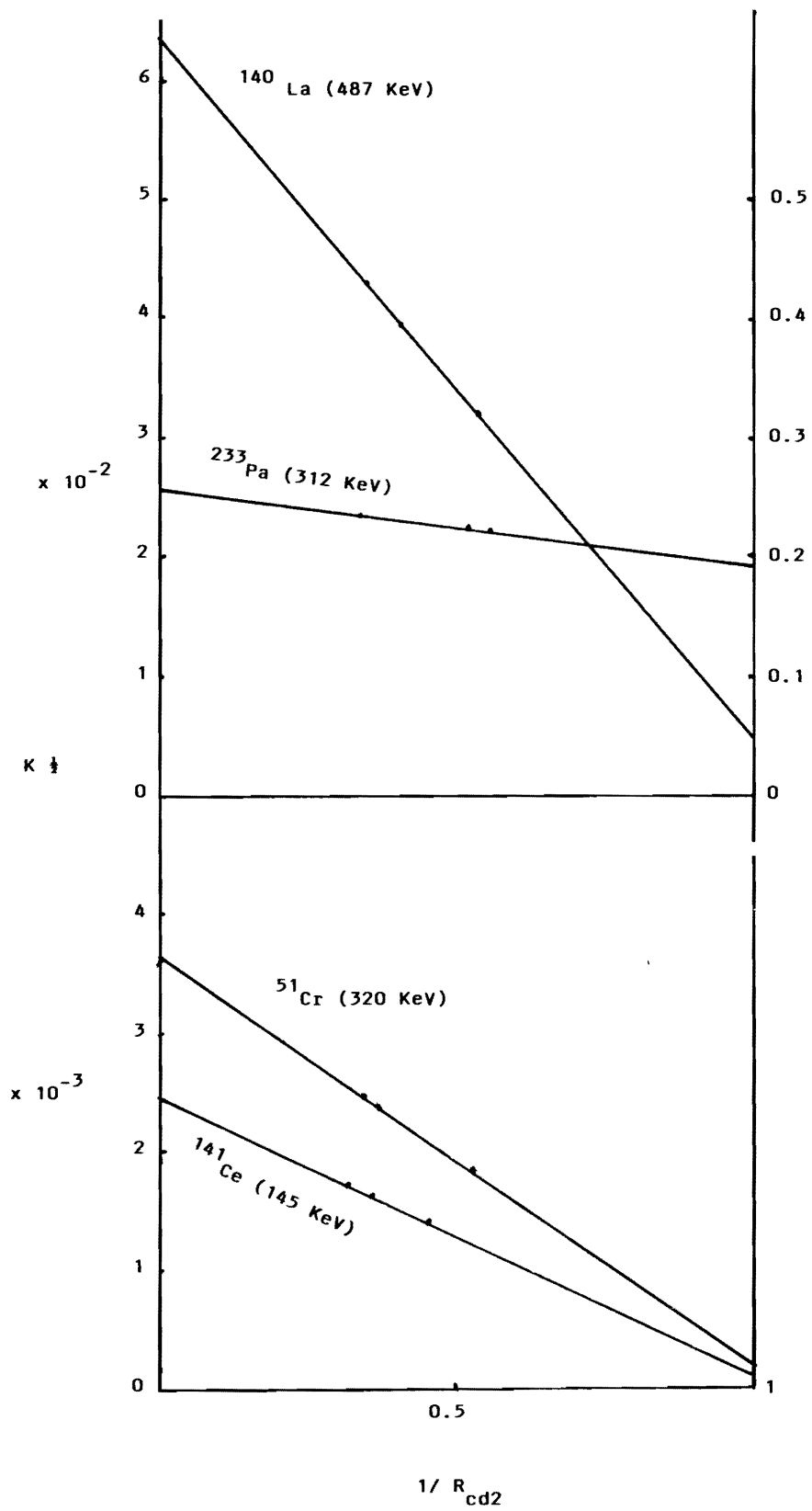
Figure 1. $K_{1/2}$ versus $1/R_{Cd2}$

Au comparators were irradiated as an Au – Al alloy and Zr as thin foils. For the element under study, use was made of materials similar to those described in reference [3,4]. Care was taken to avoid epithermal self-shielding, although sometimes small corrections had to be applied, e.g. for the 0.5% Au–Al wire (1.5% correction, Howe *et al.* 1962), and for 100 μm Zr foil (correction: 2% for ^{95}Zr and 3% for ^{97}Zr [4]. Counting was performed using a 40 cm^3 single open-ended coaxial Ge(Li) detector with a source-to-detector separation of 15 cm.

To investigate the precision and accuracy, the methods has been applied to determine the contents of elements Sc, Cr, Mn, La, Ce, Sm, and Th in USGS Standard Rock G–2, by irradiation in the Egyptian reactor.

RESULTS AND DISCUSSION

The relationship between the activation factor $K_{1/2}$ with the cadmium ratio. $1/R_{Cd2}$ of the comparator are shown in Figure 1 where $K_{1/2}$ factors were experimentally determined and calculated versus the 411.794 keV γ-line of ^{198}Au for ^{140}La (487.0 keV) ^{233}Pa (312.0 keV) ^{153}Sm (103.2 keV), ^{51}Cr (320.1 keV) and ^{141}Ce (145.4 keV) $K^0_{1/2}$ and $K^{cd}_{1/2}$ as shown in Table 1 can be obtained by extrapolating the $1/R_{Cd2} = 0$ and $1/R_{Cd2} = 1$ respectively.

The relationship between activation factors $K_{1/2}$ and $K_{3/2}$ of the pair comparator method are shown in Figures 2, 3 and 4 where $K_{1/2}$ factors were experimentally determined and calculated versus the 411.794 keV γ-line for ^{140}La (328.8, 487.0, 815.8, and

Table 1. Experimental Determination and Calculation of K_0 and $K_{e,0}$ Factors

| Isotope | E_γ (keV) | K_0 | | | $K_{e,0}$ calculated (Equation 12) |
		Theoretical (Equation 15)	Rcd – method (Equation 11)	Pair comparator (Equation 13)	
^{46}Sc	889.3	1.25		1.24	3.41×10^{-2}
	1120.5	1.25		1.25	3.41×10^{-2}
^{51}Cr	320.1	2.51×10^{-2}	2.48×10^{-3}	2.50×10^{-3}	9.13×10^{-5}
^{56}Mn	846.8	5.00×10^{-1}	—	4.99×10^{-1}	3.41×10^{-2}
^{60}Co	1173.2	1.31	—	1.31	1.71×10^{-1}
^{141}La	328.8	2.90×10^{-2}	—	2.87×10^{-2}	2.16×10^{-3}
	487.0	6.35×10^{-2}	6.38×10^{-2}	6.37×10^{-2}	4.75×10^{-3}
	815.8	3.31×10^{-2}	—	3.32×10^{-2}	2.53×10^{-3}
	1596.2	1.32×10^{-1}	—	1.34×10^{-1}	9.92×10^{-3}
^{141}Ce	145.4	3.67×10^{-3}	3.66×10^{-3}	3.65×10^{-3}	1.93×10^{-4}
^{153}Sm	103.2	2.19×10^{-1}	2.30×10^{-1}	2.29×10^{-1}	1.96×10^{-1}
^{233}Pa	312.0	2.53×10^{-2}	2.54×10^{-2}	2.51×10^{-2}	1.92×10^{-2}
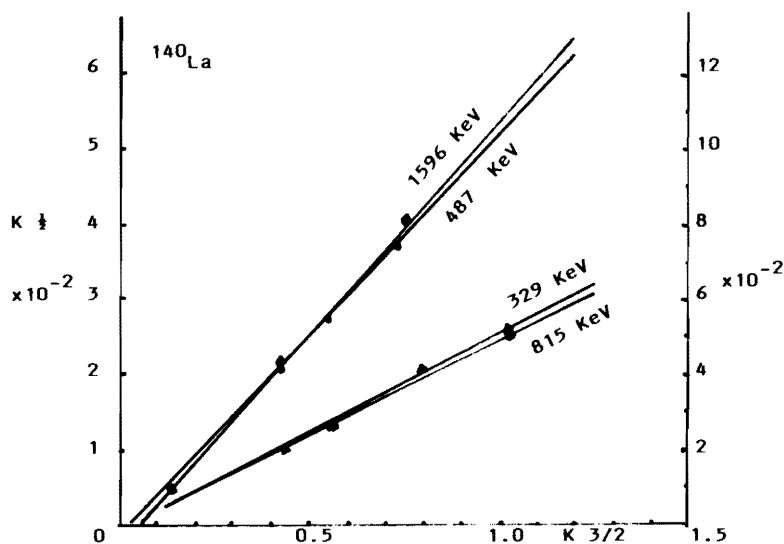
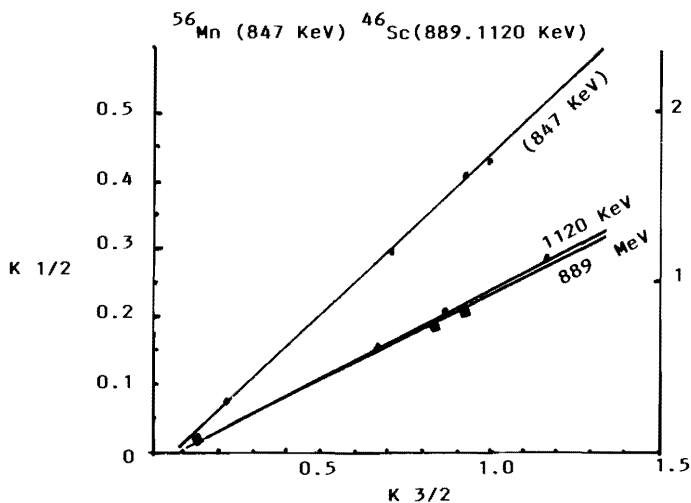*Figure 2. $K_{1/2}$ versus $K_{3/2}$ (^{140}La)*
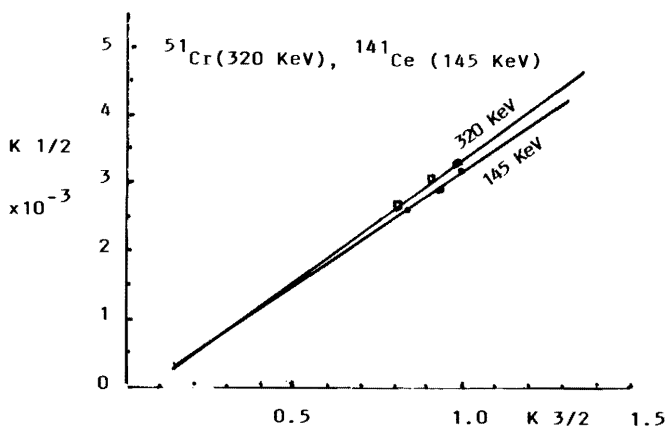
Figure 3. $K_{1/2}$ versus $K_{3/2}$ (^{56}Mn and ^{46}Sc)
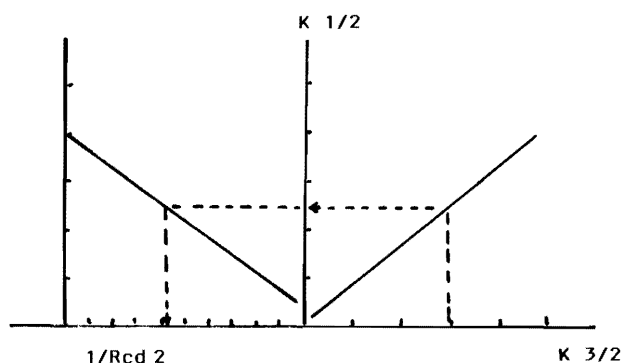

Figure 4. $K_{1/2}$ versus $K_{3/2}$ (^{51}Cr and ^{141}Ce)

Figure 5. The Relation Between $K_{1/2}$, $K_{3/2}$, and $1/R_{Cd2}$

Figure 6. $K_{1/2}$ versus $1/R_{Cd2}$
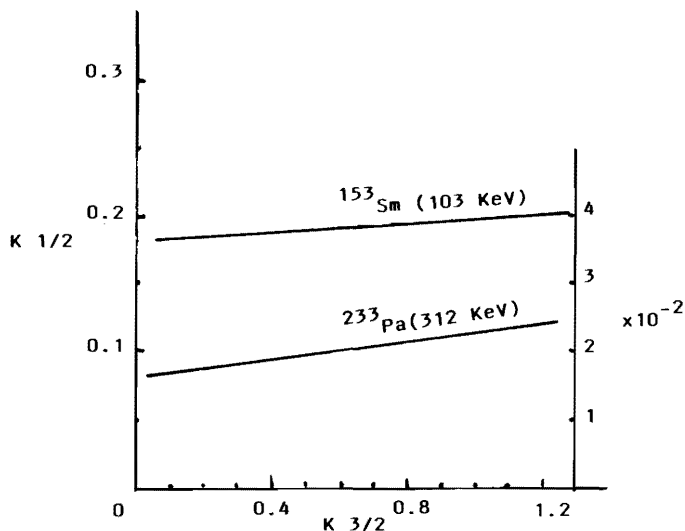
Figure 7. $K_{1/2}$ versus $K_{3/2}$ (^{153}Sm and ^{233}Pa)

1596.2 keV), ^{51}Cr (320.1 keV), ^{141}Ce (145.4 keV), ^{56}Mn (846.8 keV) and ^{46}Sc (889.3, 1120.5 keV), and the activation factor $K_{3/2}$ were experimentally determined and calculated versus the 756.7 keV γ-line of ^{95}Zr and 743.4 keV γ-line of ^{97}Zr. The relationship
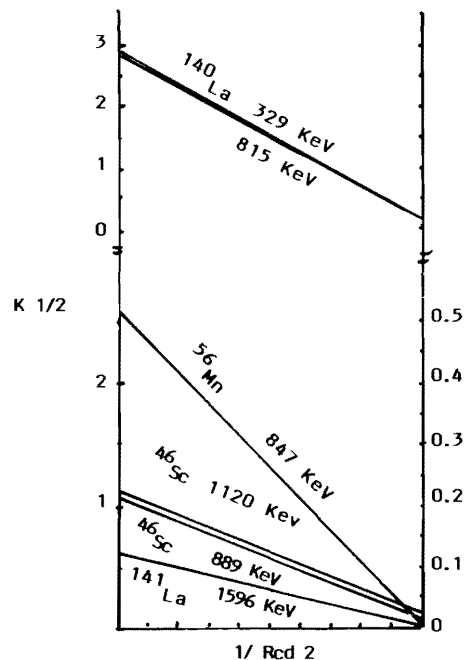
between $K_{1/2}$, $K_{3/2}$ and $1/R_{Cd2}$ according the two methods are shown in Figure 5. It is clear that the activation factors can be determined by one of the two methods as shown in Figures 6 and 7 which is based on Figure 5. In this way the two methods can be adopted and at the same time can be compared with each other. Comparison between K_0 values determined by cadmium ratio and pair comparator methods and those theoretically calculated according to equation

Table 2.   Multielement Neutron Activation Analysis of G-2

Element (ppm)	R_{Cd} – method (s%)		Pair comparator method (s%)		Reported [5]	Reported [6]
Sc	—		3.41	(1.8)	3.20	3.70
Cr	8.10	(2.2)	7.60	(2.4)	8.70	7.00
Mn(%)	—		0.0254	(0.8)	0.26	0.026
La	92.30	(3.0)	95.00	(2.3)	84.00	96.00
Ce	141.30	(2.7)	146.00	(1.9)	150.00	150.00
Sm	7.71	(0'9)	—		7.23	7.30
Th	23.20	(1.5)	—		21.80	24.20

(15) show good agreement (Table 1). The nuclear activation data and decay parameters for the isotopes concerned are adopted from the literature [3, 4]. Three samples of USGS Rock standard G–2 are analysed and the result obtained are in good agreement with the data reported in literature (Table 2).

CONCLUSION

The activation methods, namely cadmium ratio and pair comparator, presented in this work have been found to be very useful for the determinations of trace amounts of Sc, Cr, Mn, La, Ce, Sm, and Th in USGS Standard Rocks G-2. The precision and accuracy of the two activation methods used have been found to be very good. The present work has demonstrated that there are simple linear relations between the activation factors $K_{1/2}$ of different elements and $1/R_{Cd}$ of the comparator or the activation factor $K_{3/2}$ of the pair comparator, where the ratios $K_{1/2}/K_{1/3}$ are independent of spectral distribution. Therefore, it is considered a simple correction method for the problems due to the variation of the neutron spectrum, i.e. if K_0 and $K_{e,0}$ of element have been determined accurately, the $K_{1/2}$ at any irradiation position can be obtained from cadmium ratio of the comparator or $K_{1/2}$ of pair comparator in the same position. The straight line relationship between activation factors presented in this paper can be used in any other irradiation positions; this method is the most suitable one for large quantities of samples and multielement analysis without chemical separation.

ACKNOWLEDGMENT

REFERENCES

[1]   A. Simonits, F. De coste, and J. Hoste, *Journal of Radioanalytical Chemistry*, **24** (1975), p. 31.

[2]   F. Girardi, G. Guzzi, and J. Paully, *Analytical Chemistry*, **37**(1965), p. 1085.

[3]   T. Elnimr and I. I. Bondouk, *Journal of Physics D: Applied Physics*, **16** (1983), p. 1407.

[4]   T. Elnimr, A. Allian, and I. I. Bondouk, *Proceedings of the Mathematical and Physical Society of Egypt*, **58** (1984), in press.

[5]   J. C. Laul and L. A. Rancitelli, *Journal of Radioanalytical Chemistry*, **38** (1977), p. 461.

[6]   F. J. Flanagan, *Geochemica et Cosmochimica Acta*, **33** (1969), p. 80.